ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

Дубна

E7-98-178

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SEARCH FOR SPONTANEOUS FISSION OF ^{226}Ra AND SYSTEMATICS OF THE SPONTANEOUS FISSION, α-DECAY AND CLUSTER DECAY PROBABILITIES

Submitted to «Ядерная физика»

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1998

1. Introduction

The discovery of cluster radioactivity of radium isotopes and more heavy nuclides gave new reasons for investigation of coupling between different channels for spontaneous emission of multinucleon particles by atomic nuclei.

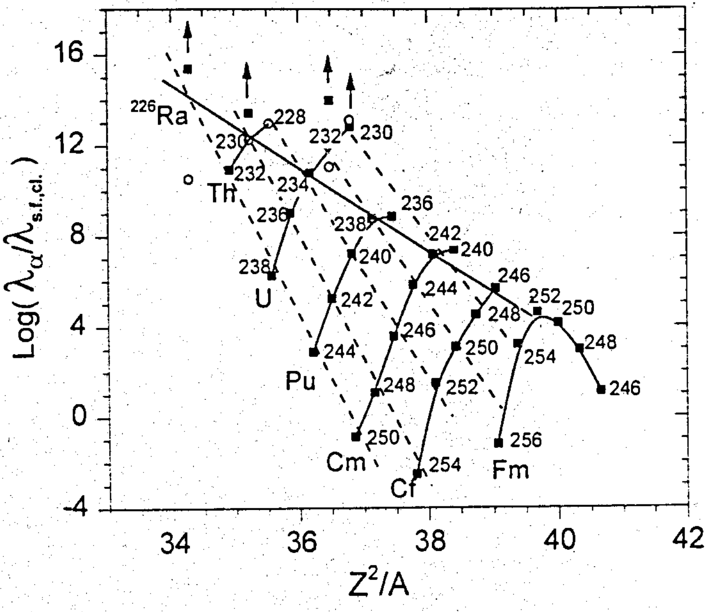

Fig. 1. The dependence of the ratios of the α-decay and spontaneous fission probabilities (■) and α-decay and cluster decay probabilities (o) on the fissility parameter Z^2/A. The dashed lines connect the points corresponding to $\Delta Z=2$ and $\Delta A=6$. For the nuclear chain including ^{226}Ra the straight line connecting the points for $\Delta Z=2$ and $\Delta A=4$ is drawn.

The correlations between the α-decay and spontaneous fission probabilities were found shortly after accumulation of the experimental data on properties of transuranium nuclides [1]. The corresponding systematics is shown in Fig.1. For the nuclei with atomic numbers $Z \le 92$ the cluster decay probability is comparable or higher than the spontaneous fission probability [2]. The systematics connecting the spontaneous fission and α-decay probabilities can be expanded for the sum of the spontaneous fission and cluster decay probabilities [3,4]. A question arises now about possible coupling of the spontaneous fission and cluster decay channels. Accordingly, it is desirable to have the data on the spontaneous fission of nuclei with $Z \le 90$. Such nuclei have fission barriers of a complicated form for which theoretical calculations show more than two maxima [5,6]. The calculations of spontaneous fission probabilities for nuclei in this domain are not reliable. Due to that it is advisable to investigate experimentally spontaneous fission of nuclei with $Z \le 90$. ^{232}Th is the lightest nucleus for which spontaneous fission was observed [3]. The measured value of the ^{232}Th spontaneous fission probability fits well with the systematics of Fig.1. The highest limit for spontaneous fission was measured for natural lead. It is equal to $T_{1/2\,s.f.} > 10^{25}$ years [7]. The measured up to now limit of ^{226}Ra spontaneous fission corresponds to $T_{1/2s.f.} > 10^{14}$ years [8]. From the systematics shown in Fig.1 the partial half-life value for the ^{226}Ra spontaneous fission equal to 10^{17}-10^{18} years can be expected. It is much less than the extrapolations from other systematics. According to the systematics connecting the values of $T_{1/2s.f.}$ with the fissility parameter Z^2/A and including corrections for the deviations of experimental nuclear masses from the corresponding liquid drop values [9,10] the half-life for the ^{226}Ra spontaneous fission can be $\approx 10^{25}$ years.

The goals of presented work are: 1) search for the ^{226}Ra spontaneous fission with the sensitivity allowing one to specify the limits of using the systematics shown in Fig.1; and 2), revealing of physical grounds of this systematics.

2. Experiment

A ^{226}Ra source with an intensity of 2mCi (7.4×10^7 α/s ^{226}Ra) has been prepared at the Institute of Nuclear Physics (Orsay, France) by the selfprecipitation method. It is a $RaSO_4$ layer put in the form of circle 15 mm in diameter on a platinum backing. Special phosphate glasses made at the St. Petersburg Optical Institute (Russia) have been used for detecting the fission fragments. The size of the glasses was 40×50 mm^2. The exposition of the detectors has been performed in air in an isolated box preventing contamination of the environment by radon. The distance between the surfaces of the glass detectors and the ^{226}Ra source was 1 mm. The ^{226}Ra spontaneous fission fragments were to registered in a circle 16 mm in diameter in the center of the phosphate glass. The remaining surface of the glass served for the background measurements. The fission fragment registration efficiency was 0.8 in a solid angle of 2π. The calibration of detectors was performed by the ^{248}Cm spontaneous fission fragments through an orifice of ≤1 mm in diameter in polyethylenetherephthalate plastic film protecting the rest of the glass surface during the calibration. New plastic film was used for every calibration to prevent contamination of the detectors by microquantities of ^{248}Cm. It was found that starting from the fluence of $\approx 2\times10^{14}$ α/cm^2 the diameters of the fission fragments tracks, observed with an optical microscope, grow in size after similar etching in HF acid in comparison with the tracks on the non-irradiated by α-particles phosphat glass surface.

We have performed 6 expositions of glass detectors on the ^{226}Ra source with the total duration of 660 days. In the expositions lasting from 5 to 316 days, the α-particle fluences from the decay of ^{226} Ra and its daughter products (^{222}Rn, ^{218}Po, ^{214}Po, ^{210}Po) varied from 3.2×10^{13} up to 2.0×10^{15} α/cm^2. After all the expositions we have registered one fission fragment track in the central region 16 mm in diameter and three tracks at all the remaining glass surfaces. The ratio between the central and other surfaces of the glass detectors used for the background estimation is 1:9. The statistically significant difference between a possible effect and background is absent. It follows from our measurements that for ^{226}Ra the ratio between the α-decay and spontaneous fission probabilities $\lambda_\alpha/\lambda_{s.f.} \geq 2.6\times10^{15}$. Correspondingly, $T_{1/2\ s.f.} \geq 4\times10^{18}$ years at the confidence level of 63% for the Poisson distribution of events in time. The value of the limit for the ratio $\lambda_\alpha/\lambda_{s.f.}$ obtained for ^{226}Ra can be used with an accuracy of the factor ≈2 for its daughter ^{222}Rn which in ≈50% of cases of the ^{226}Ra α-decays implants into a platinum backing due to a recoil impulse.

3. Analysis of the results

The obtained limit for the ^{226}Ra spontaneous fission probability proved to be ≈50 times less than that expected from the systematics in Fig.1. A similar situation exists for the properties of ^{232}U, ^{230}Th, and ^{228}Th. The measurements performed in the work [11] have shown that the effect assigned in previous works to spontaneous fission is in reality emission of ^{24}Ne clusters with the energy of 55.9 MeV. The limit for the ^{232}U spontaneous fission probability measured in work [11] is of three orders of magnitude less than that expected from the systematics in Fig.1. It was mentioned in works [3,4] that the deviations from the systematics in Fig.1 can be caused by the competition between the channels of spontaneous fission and cluster decay. The cluster decay probabilities of ^{232}U, ^{228}Th, and ^{230}Th are higher than probabilities of non observed up to now spontaneous fission. The corresponding points for $\lambda_\alpha/\lambda_{cl}$ fit well with the points for $\lambda_\alpha/\lambda_{s.f.}$ of other nuclei in the systematics presented in Fig.1. For the ^{14}C cluster decay of ^{226}Ra $\lambda_\alpha/\lambda_{cl}=(3.3\pm1.0)\times10^{10}$ [12-14]]. The corresponding point is presented in Fig. 1. Apparently, the relatively high cluster decay probability of ^{226}Ra can suppress its spontaneous fission.

Alpha-decay, spontaneous fission and cluster decay are quantum transitions from one initial nucleus to two other nuclei. The term "quantum" means that the initial and final experimentally observed nuclear states are separated by an energy-space interval which cannot be experimentally investigated. From this point of view, one can try to use theoretical fission models statistically considering relative probabilities of fission fragment formation as a function of their potential energy near the scission point [15-18] for cluster decay and α-decay with the aim of analysis of the competition between these three modes of nuclear decay. In the first approximation the relative yields of products of the two-body nuclear decay can be described by the equation:

$$\mathbf{W=const\times exp[-(Vc-Q)/T]} \qquad (1),$$

where V_c is the Coulomb energy of two touching final nuclei with the atomic numbers Z_1 and Z_2 and mass numbers A_1 and A_2. Q is the ground state mass difference of the initial and the final nuclei. T is the parameter expressed in energy units. T can be considered as a nuclear temperature or parameter inverse to the nuclear level density parameter at $(V_c\text{-}Q)<0$. At $(V_c\text{-}Q)>0$ one can consider T as the energy of zero nuclear vibrations, which used, for example, in the formula for parabolic fission barrier penetrability [19]. Formula (1) is in fact the Boltzmann distribution and it means that for a system of nucleons of the initial nucleus transitions to the nuclear states with the minimal potential energy are more probable. In our case the potential energy is the sum of Coulomb energy and nuclear potential energy presented by the Q-value.

In Fig.2 the partial half-lives for spontaneous fission, α-decay and cluster decay of nuclei presented in Fig.1 are shown as functions of the corresponding $(V_c\text{-}Q)$-values. The experimental data on α-decay, cluster decay and spontaneous fission are taken from works [20], [21] and [22], correspondingly. We consider the properties of the even-even nuclei to exclude hindrance factors manifesting themselves in the odd-nuclei decay. Only in the case of the cluster decay we have presented the data for odd nuclides due to a relatively small number of known cluster emitters. Really, including the data for odd nuclei results a some increase in the point dispersion but does not change the general features of the experimental data systematization in Fig.2. The V_c-value was calculated according to:

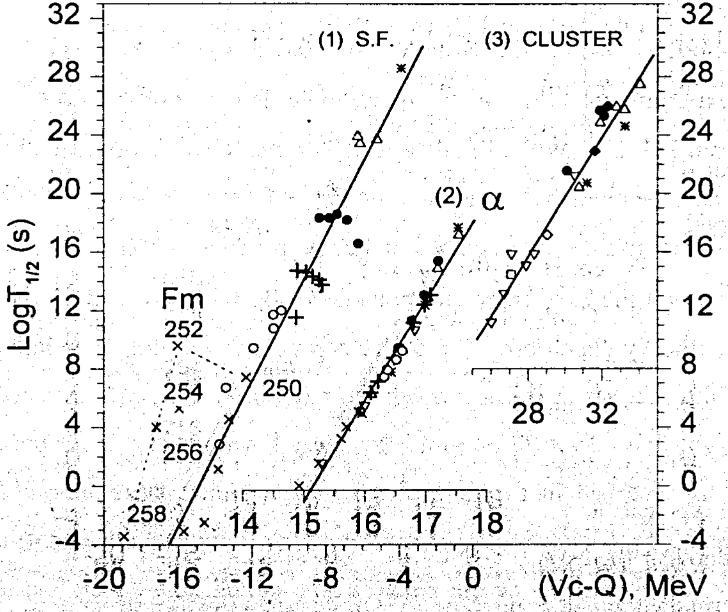

Fig. 2. The dependences of decimal logarithms of the partial half-lives for spontaneous fission (1), α-decay (2) and cluster decay (3) on the corresponding values of $(V_c\text{-}Q)$, i.e., difference between Coulomb energy near the scission point and decay energy.(∇)-Ra isotopes, (*) – Th, (Δ)-U, (●)-Pu, (+)-Cm, (o)-Cf, (x)-Fm, (□)-Fr, (◊)-Ac, (◆)-Pa.

$$\mathbf{Vc=1.44\times(Z_1\times Z_2/R)\times F,\ MeV} \qquad \mathbf{(2)}$$

For α-decay the distance between the centers of nuclear charge distributions is:

$$R=1.44\times[4^{1/3}+A_2^{1/3}\times(1+0.633\times\beta_2)],\ \text{fm};$$

for cluster decay:

$R=1.44\times(A_1^{1/3}+A_2^{1/3})$, fm;

for spontaneous fission:

$R=1.44\times[A_1^{1/3}\times(1+0.633\times\beta_2)_1+A_2^{1/3}\times(1+0.633\times\beta_2)_2]$, fm.

The experimental values of the quadrupole deformation parameters β_2 were taken from [23, 24]. When the data for some nuclei were not available in [23, 24] we used the empirical regional systematics from [25]. We approximated the forms of the fission fragments and heavy products of α-decay by prolate spheroids with the axes calculated for known β_2 values according to [26]. For the cluster decay the deformation of decay products was not taken into account. Taking into account the cluster quadrupole deformations leads to a splitting of one straight line into different lines for every Z values of the clusters. Similar splitting is observed in the generalized Geiger-Nuttal systematics [27]. It is explained on the basis of the spectroscopic factors for the cluster formation [2, 28]. The systematization of the whole experimental data on the cluster decay using one straight line in Fig.2 shows the equivalence of accounting for the influence of the cluster properties on the cluster emission probability with either a spectroscopic factor or deformation (the heavy cluster decay product is the almost spherical nucleus ^{208}Pb or its neighbour).

For the Q-value calculations we used experimental nuclear masses from [29]. When the experimental data for some nuclei were not available we used the masses calculated according to mass formulae giving the best fit to the closest experimental data in this nuclear region [30]. The factor F characterizes deviation of Coulomb interaction of touched coaxial spheroids from the point charge interaction $(Z_1\times Z_2)/R$ [31]. The use of the experimental values of the nuclear masses and deformations makes the potential energy calculations much more simple and reliable in comparison with the model consideration of a consecutive changes of the form and energy of a fissioning nucleus. The variations of the radius parameter r_0 from 1.2 fm to 1.7 fm have shown that r_0=1.44 fm provides the least scattering of points relative to the straight lines in Fig. 2. For spontaneous fission we have used the minimal $(V_c\text{-}Q)$-values from all the variants of charge and mass ratios at double splitting of an initial nucleus. So, shown in Fig.2 the $(V_c\text{-}Q)_{s.f.}$-values correspond to the maxima of the fission fragment yields. It is illustrated in Fig.3 where the change of the potential energy in the ^{226}Ra decay into two fragments is shown as a function of the value of atomic number Z_1 of a more light two-body decay product. Comparison of the curves obtained without and with taking into account the ground state nuclear quadrupole deformations shows their essential role in formation of the charge and mass distributions of two-body decay products.

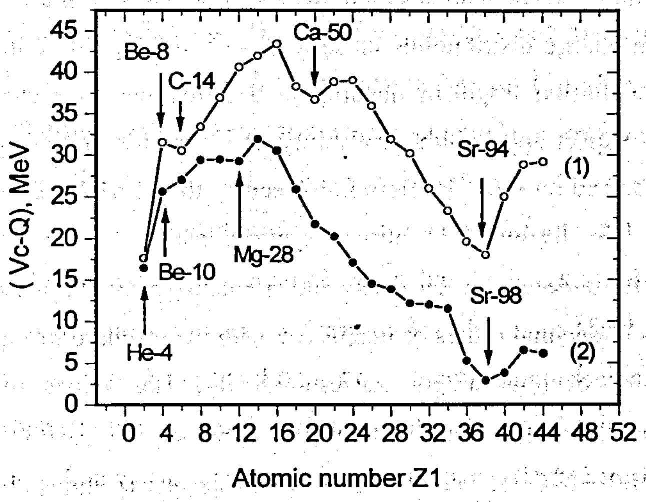

Fig.3. The dependence of potential energy $(V_c\text{-}Q)$ of the ^{226}Ra two-body decay on the atomic number Z_1 of a more light product. (1)-without taking into account deformations of the decay products. (2) - with taking into account the ground state quadrupole deformations of final nuclei.

Without taking into account the product deformations, one can practically expect only asymmetric spontaneous (and low exitation energy) fission of ^{226}Ra with maxima in charge distributions close to $Z_1=38$ and $Z_2=50$. With taking into account the fission fragment quadrupole deformations (see curve 2 in Fig.3), one can expect appreciable probability of symmetric fission of ^{226}Ra. This is what observed for the ^{226}Ra fission induced by the 11 MeV protons [32].

Fig.2 shows that formula (1) allows systematization of the half-lives ($T_{1/2}$=const/W) for α-decay, cluster decay and spontaneous fission changing in the range of 20-30 decimal orders of magnitude with the standard deviations of 0.7, 1.2, and 2.0 decimal orders, correspondingly. The values of the T parameter determined by the least-squares method are (0.064±0.003), (0.21±0.02), and (0.18±0.01) MeV for α-decay, cluster decay and spontaneous fission, correspondingly. The points for the Fm isotopes with the neutron number N≥152 were not taken into account at the determination of the T parameter for the spontaneous fission. Analysis of deviations from the systematics in Fig. 2 connected with the higher stability of nuclei in neighbourhood of neutron closures N=126, 152 and 162 (first of all with regard to spontaneous fission) is beyond the scope of the present work. Preliminary analysis shows that for taking into account the influence of neutron closures it is necessary to increase the T-value in the vicinity of N=152 and to decrease it in the vicinity of N=140-142 between neutron closures N=126 and N=152. These conclusions are consistent with the changing of a nuclear rigidity coefficient at the quadrupole vibrations [33] and a nuclear level density parameter [34] as a function of the neutron number.

The ratio of the α-decay and spontaneous fission probabilities when using systematics in Fig.2 can be approximated for the nuclei with Z≤98 by the relation:

$$\mathrm{Log}(\lambda_\alpha/\lambda_{s.f.})=\{140+[-(V_c-Q)_\alpha/0.064+(V_c-Q)_{s.f.}/0.18]/2.3\} \qquad (3)$$

For the ratio of the α-decay and cluster decay probabilities:

$$\mathrm{Log}(\lambda_\alpha/\lambda_{cl})=\{60.5+[(V_c-Q)_\alpha/0.064+(V_c-Q)_{cl}/0.21]/2.3\} \qquad (4)$$

The numerical coefficient 2.3 transferring natural logarithm to decimal ones has been distinguished to use the same T-values in the formulas (1), (3) and (4).

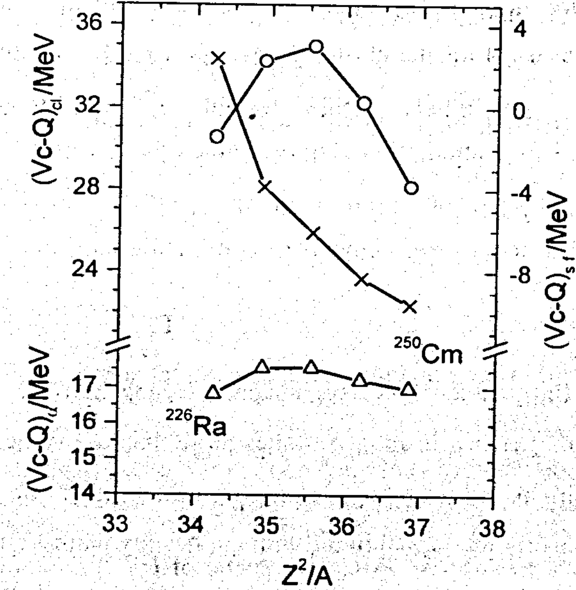

Fig. 4. The dependence of the potential energy (V_c-Q) near the scission point for α-decay (Δ), spontaneous fission (x) and cluster decay (o) of ^{226}Ra (cluster ^{14}C), ^{232}Th (^{26}Ne), ^{238}U (^{34}Si), ^{244}Pu (^{46}Ar), ^{250}Cm (^{46}Ar) on the fissility parameter Z^2/A.

From the data in Fig. 2 and formula (3) it follows that linear dependence of $\mathrm{Log}(\lambda_\alpha/\lambda_{s.f.})$ on Z^2/A for the nuclear chains with ΔZ=2 and ΔA=6 or 4 in Fig 1 must be the result of similarity of the $[(V_c-Q)/T]_\alpha$ and $[(Vc-Q)/T]_{s.f.}$.

dependence on Z^2/A. Fig. 4 shows the $(V_c\text{-}Q)$-values for the spontaneous fission, cluster decay and α-decay of nuclei in the chain with $\Delta Z=2$ and $\Delta A=6$ in Fig. 1 including ^{226}Ra. The linear Z^2/A dependence of $\mathrm{Log}(\lambda_\alpha/\lambda_{s.f.})$ for ^{250}Cm. ^{244}Pu, ^{238}U, ^{232}Th in Fig. 1 correlate with the similarity of Z^2/A dependences of $(V_c\text{-}Q)$ for α-decay and spontaneous fission in Fig. 4. The ^{226}Ra $(Vc\text{-}Q)_\alpha$-value is decreasing sharply in comparison with a monotonous character of changing these values for more heavy nuclei in the same nuclide chain. The same behaviour is observed for the cluster decay. But $(V_c\text{-}Q)_{s.f.}$ continues to change monotonically. Accordingly, using formula (3) we obtain for ^{226}Ra $\mathrm{Log}(\lambda_\alpha/\lambda_{s.f.})= 31$ in contradiction with the value of ≈13.5 as it follows from the purely empirical systematics in Fig. 1. So, the absence of the ^{226}Ra spontaneous fission fragments in our measurements becomes undestandable.

4. Conclusions

1) The low limit of the ^{226}Ra spontaneous fission half-life corresponds to $T_{1/2}\geq 4\times 10^{18}$ years. It is about 50 times more than the value expected from the empirical systematics connecting the ratios of α-decay and spontaneous fission probabilities with the fissility parameter Z^2/A [1, 3].

2) The α-decay, cluster decay and spontaneous fission probabilities of nuclei with $88\leq Z\leq 100$ can be systematized as functions of the corresponding values $(V_c\text{-}Q)$- difference between Coulomb energy near the scission point of final decay products and decay energy Q. The experimental values of nuclear masses and quadrupole deformations are used for the $(V_c\text{-}Q)$ calculations.

3) The systematics of $\mathrm{Log}(\lambda_\alpha/\lambda_{s.f.})$ as a function of Z^2/A (Fig 1 and refs. [1, 3]) is the consequence of similarity of the Z^2/A dependences of the $(V_c\text{-}Q)_\alpha$ and $(Vc\text{-}Q)_{s.f.}$ values for nuclear chains with $\Delta Z=2$ and $\Delta A=6$ or 4. In the case of ^{226}Ra as in the case of ^{232}U the proportionality between $\mathrm{Log}(\lambda_\alpha/\lambda_{s.f.})$ and Z^2/A for the nuclear chains with $\Delta Z=2$ and $\Delta A=6$ (see Fig. 1) is destroyed due to disruption in similarity of the Z^2/A dependences of $(V_c\text{-}Q)_\alpha$ and $(V_c\text{-}Q)_{s.f.}$. According to the presented in this work systematics (Fig. 2) the expected spontaneous fission half-life for ^{226}Ra is $\approx 10^{34}$ years and for ^{232}U $\approx 10^{21}$ years. But the cluster decay probabilities for both these nuclei are close to the values, expected for spontaneous fission according to systematics in Fig. 1.

This work has been supported by the Russian fund of the fundamental investigations (Grant No 96-02-17975.)

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Received by Publishing Department
on June 19, 1998.